PLANT TRANSFER FACTORS FOR PLUTONIUM AND AMERICIUM AT ROCKY FLATS: A REVIEW AND ANALYSIS

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September 23, 2001

Introduction

This report was prepared at the request of the Rocky Flats Citizen's Advisory Board. It's purpose is to provide a review and analysis of the development of soil to plant transfer parameters that can be used in RESRAD computations to support an independent assessment of residual soil action levels for plutonium and americium in surface soil at Rocky Flats. My review is based primarily on historical field research which I directed at Rocky Flats and laboratory experiments at Colorado State University, using soil and vegetation sampled from Rocky Flats. This work was conducted between 1982 and 1995. In addition, a 1992 field study designed to specifically measure plant/soil concentration ratios (CRs) at a site at the Savannah River Site (SRS) in South Carolina was deemed relevant to this effort because this study site was not influenced by resuspension, and yet the levels of $^{239,240}Pu$ and ^{241}Am were sufficient to permit accurate measurements of these radionuclides in crop plants and in the soils the crops were growing in. Because the SRS soils were sandy and acidic, the plant uptake data would likely represent a very conservative CR estimate for Rocky Flats, for the true root uptake pathway. Of course, the additional pathways of aerial resuspension from wind and rainsplash operate at Rocky Flats, so field and laboratory data from our work at Rocky Flats is important, because it can be used to help quantify the various pathways of plant contamination and their relative importance.

This report is organized around six specific questions asked of me by the Residual Soil Action Level (RSAL) working group. These questions are appended to this report.

Responses to Questions Asked by the RSAL Working Group

1. **Plant/Soil Transfer Factor:**

The plant/soil transfer values (which I will call plant/soil concentration ratios, or CRs) listed in a document provided by the RSAL working group are similar to those which can be derived from the SRS study (Whicker et al., 1999). The geometric mean values from the SRS study were, for leafy vegetables (turnip greens): 2.3 E-03 for Pu and 5.3 E-02 for Am. The geometric mean values for non-leafy vegetables (bush beans, corn kernels, and turnip tuber) were 1.9 E-04 and 4.5 E-03 for Pu and Am, respectively. These values are based on a dry plant mass and dry soil mass basis, and reflect true root uptake only. The values could be changed to a wet plant and dry soil basis using the Baes dry mass/wet mass ratios as proposed by the RSAL working group. I am not certain whether RESRAD 6.0 treats this factor as true root uptake or as plant/soil ratios resulting from both root uptake and aerial resuspension pathways. If the former, I think these values could be used as credible, yet reasonably conservative values for plant transfer, provided of course that RESRAD 6.0 also treats resuspension separately. If however, RESRAD 6.0 treats this parameter as one which results from all these pathways, then the values from Whicker et al. (1999) would not be high enough.

In the latter case, the plant/soil CR values should reflect all pathways. Historical plant and soil measurements in the field at Rocky Flats by Little et al. (1980) and by Webb et al. (1993) are relevant because the soil and vegetation samples were taken in the same locations (CSU Macroplot 1), and the vegetation measurements reflected all transport pathways because the vegetation samples were only clipped, air-dried, and ground prior to analysis. They were not washed, so radioactivity in surficial dust from aerial resuspension pathways would be inherently included, in addition to root uptake. Soil concentrations were also measured as a function of depth in the soil, so the data can be used to estimate an average soil concentration for various soil sampling depths. For example, by integrating the Macroplot 1 soil concentration versus depth function obtained for Rocky Flats soil in 1989 by Webb et al. (1993), one can obtain a function for the mean Macroplot 1 soil concentration $\overline{C}_{soil}$ as a function of the depth from the surface (d, in cm) of a soil sample:

$$\overline{C}_{soil} = \frac{3.4x10^4 Bq \cdot cm / kg}{d}\left(1 - e^{-0.233d}\right)$$

Using this relationship and the data in Little et al. (1980) and in Webb et al. (1993), one can calculate for a soil sample taken from 0-15 cm for example, that the field plant/soil CR for Pu in 1973 was ~ 2.0 E-01, and in 1989 was ~6.2 E-02. This calculation indicates two major points, namely that resuspension is the primary mechanism of plant contamination by Pu at Rocky Flats, and that resuspension is declining over time. These phenomena have been shown by many other investigators, but this, to my knowledge, is the only relevant site-specific data for Rocky Flats. Based on this calculation for 0-15 cm soil concentrations, I would recommend a conservative CR value that reflects all pathways as 6 E-02 for Pu. The current value may actually be 2 or 3 times lower, but this is only speculative, as we have no recent data. Furthermore, if the land is disturbed in a way that increases local resuspension, then a more conservative figure (0.2 to 6 E-02) seems justified.

We do not have direct measurements for Am in vegetation, so if a CR value reflecting all plant contamination pathways is used in RESRAD 6.0, then I would recommend using the same value as for Pu. The reason for this is that most of the contamination on plants in the field situation results from aerial resuspension. The solubility differences for Pu and Am would only affect the root uptake pathway. Again, this approach is likely on the conservative side, but the lack of Am data for plants at Rocky Flats suggests this to be a reasonably cautious assumption.

2. **Rooting Depth for a Vegetable Garden**

I believe that the assumption of 15 cm for the rooting depth of a vegetable garden is reasonable. However, the mean Pu or Am in the soil which is used to drive the RESRAD calculations is very sensitive to the depth of the sample used to estimate the actual concentration in Rocky Flats soil (Webb et al., 1993; Hulse et al., 1999). Therefore, the soil data used to drive a calculation, or a residual soil action level that implies a given dose to a resident farmer, must clearly specify the soil depth and correct for it, as I did in the calculation above.

3. **Assumption of Contaminant Homogeneity in Soil**

Clearly, as noted above, if the soil sample used to derive the dose or action level calculations was taken from the upper ¼ inch, or the upper 2 inches, then the CR values applied may not be appropriate, unless they were derived for this particular depth, as shown in my answer to question 1, above. With regard to spatial inhomogeneity that may occur over and above the rather predictable variations with depth (Hulse et al., 1999), I have not seen evidence that this is a serious problem. Simon and Fraley (1986) investigated this problem for ^{226}Ra uptake in sagebrush and concluded that the average uptake by plants was independent of the degree of homogeneity in the soil. However, these scientists did find that there was less variability among individual plants and leaves in uptake if the soil was uniformly contaminated. There is considerable small-scale (centimeters or less) variability in Pu concentrations in Rocky Flats soils, but this should be averaged out for applications to larger areas, such as a garden for example.

4. **Foliar Deposition Versus Root Uptake**

A study by Jarvis (1991) is relevant to estimating root uptake of Pu by plants from Rocky Flats soil. Jarvis used soil taken from CSU macroplot 1 at Rocky Flats and prepared plant growing pots in the CSU greenhouse with this soil. After seeding, the pots were covered with uncontaminated sand to prevent resuspension and surficial contamination. Jarvis found that true root uptake was extremely small, with CR values in the range of ~3 E-05. Uptake in plants growing on soil cores that were not covered with sand, thus permitting resuspension, were on the order of 2 E-03. This is additional evidence that aerial transport pathways dominate the accumulation of Pu on plants at Rocky Flats, and that root uptake can be essentially ignored in predictive models for this radionuclide. Root uptake of course should always be evaluated and included in models for the sake of credibility and completeness, but this pathway is not likely to make a significant difference for Pu in the overall outcome for Rocky Flats. For Am on the other hand, root uptake may be relatively more important.

We at CSU did not conduct any studies at Rocky Flats where we tried to directly measure foliar deposition. Nevertheless, if reasonable assumptions are made with respect to loss rates of surficial materials from plants, one can use the data of Little et al. (1980) and Webb et al. (1993) to estimate foliar deposition rates. This would be a relatively straight-forward exercise.

5. **Estimating Rainsplash on Plants**

We used an experiment by Dreicer et al. (1984) to estimate a rainsplash-driven soil to plant transfer rate constant of 8.6 E-04 d^{-1} in the PATHWAY foodchain model (Whicker and Kirchner, 1987). This was based on experimental data and simple model calculations. We also derived from the product of a resuspension factor (10^{-5} m^{-1}) and a deposition velocity (173 m d^{-1}) an aerial resuspension rate constant of 1.7 E-03 d^{-1} in the PATHWAY code (Whicker and Kirchner, 1987). This would indicate a ratio of aerial deposition to rainsplash of about 2. Of course, these parameters are uncertain and will vary greatly over space and over time intervals that are relatively short (days-weeks). Time-averaged variations in these parameters, however, become much smaller. Nevertheless, the PATHWAY code has been shown in extensive validation testing exercises to predict independently measured radionuclide concentrations in plants with quite reasonable accuracy (Kirchner and Whicker, 1984), suggesting that the parameters cited above produce reasonably accurate simulations of reality.

If the aerial deposition pathway parameters were estimated using the approach of combining loss rate estimates with actual field data from Rocky Flats, as mentioned in my response to question 4, the aerial soil to plant transfer parameter thus estimated would inherently include both the processes of dry resuspension/deposition and rainsplash. This exercise is recommended if RESRAD 6.0 specifically includes the aerial deposition process.

6. **Time-Dependence of Plant/Soil CR Values**

As discussed on page 2, 3rd paragraph, in my response to question 1, there is clearly a time dependence, namely a temporal decline, of field CR values in Rocky Flats vegetation. The basic reasons for this decline are that initial resuspension of a fresh deposit depletes the source of easily-resuspendable material; some contamination on the soil surface can percolate deeper into the soil by various physical, chemical and biological mechanisms; and deposition of normal, uncontaminated dust over time helps cover and protect deeper layers of material from resuspension. In range-type ecosystems where the soil is not mechanically disturbed to a significant degree, as from tilling, these processes clearly operate to reduce resuspension over time, as shown by Anspaugh et al. (1975) and numerous other researchers.

On the other hand, I would expect that in a tilled agricultural situation, this sort of temporal decline may not occur because the periodic mechanical mixing resulting from plowing would negate the processes described above by periodically bringing sub-surface soil back to the surface. I am not aware of actual long-term measurements in tilled fields that show this, however. Lacking data on this question for deposits on tilled fields, it is prudent to assume that temporal declines may not occur.

The effect of tilling a field having an initial deposit on the surface is described in the PATHWAY model (Whicker and Kirchner, 1987).

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